

EXECUTIVE SUMMARY

I. INTRODUCTION

This report has been prepared to summarize the findings of the Y-12 National Security Complex (Y-12 Complex) Mass Balance Project and to support preparation of associated U. S. Department of Energy (DOE) site reports. The project was conducted in support of DOE efforts to assess the potential for health and environmental issues resulting from the presence of transuranic (TRU) elements and fission products in recycled uranium (RU) processed by DOE and its predecessor agencies. The United States government used uranium in fission reactors to produce plutonium and tritium for nuclear weapons production. Because uranium was considered scarce relative to demand when these operations began almost 50 years ago, the spent fuel from U.S. fission reactors was processed to recover uranium for recycling.

Uranium that has been irradiated in reactors contains TRU elements [e.g., plutonium (Pu) and neptunium-237 (Np)], fission products [e.g., technetium-99 (Tc)], and reactor-generated uranium products [e.g., uranium-236 (^{236}U)]. Following chemical processing to extract various isotopes of Pu and tritium, as well as to recover uranium for reuse, trace quantities of Pu, Np, Tc, and ^{236}U remain in the RU stream. These constituents make the RU stream more radioactive than natural uranium. Thus, the processing and re-enrichment of RU may present an increased potential for personnel and environmental exposure greater than that normally associated with the processing of unirradiated uranium.

In response to these concerns, DOE initiated an effort to identify all situations in which the processing of RU by DOE and its predecessor agencies could have created an increased potential for exposure of workers and/or significant increased environmental exposure. The first step in this process involves the “mass balance review.”

The Y-12 Complex Mass Balance Project represents an effort to collect, verify, analyze, and interpret available data to provide an overall accounting, or site mass balance, for Y-12 Complex RU streams. In addition, data on related Y-12 Complex processes and activities and data on Pu, Np, Tc, and ^{236}U —the primary constituents of concern in the RU streams—have also been collected, analyzed, and interpreted. Based on available Y-12 Complex records and information about processes and methods of operation and maintenance, the Project Team has identified essentially all those plant activities that (1) created a likelihood of Y-12 Complex workers coming into contact with significant levels of RU constituents through direct physical contact or via airborne dust and/or (2) caused reportable environmental releases of concentrated RU constituents.

The Project Team analyzed data on receipts, shipments, inventories, product, releases, and other categories—along with available analytical data—in the context of documented historical information on Y-12 Complex processes and activities. Understanding of processes known to concentrate Pu, Np, and Tc and of activities known to create potential for exposure to these RU constituents provided additional context for analysis. By correlating mass balance data, analytical data, and historical information on Y-12 Complex processes, the team was able to identify specific processes, locations, and time periods of importance for potential worker exposure or environmental releases. These processes, locations, and time

periods became the focus of additional assessment to determine the situations that had the potential to create exposure hazards for workers and/or significant environmental release.

II. CHARACTERIZATION OF RU STREAMS RECEIVED AT THE Y-12 COMPLEX

Uranium streams received at the Y-12 Complex that contained or may have contained RU constituents included:

- highly enriched RU material in the form of uranyl nitrate (UN) solutions or uranium oxide (UO₃) received from the Savannah River Site (SRS) and the Idaho Chemical Processing Plant (ICPP) and
- slightly depleted RU¹ oxide (including ash and scrap) from the Oak Ridge Gaseous Diffusion Plant (ORGDP), Hanford, and the Paducah Gaseous Diffusion Plant (PGDP).

In accordance with the methodology prescribed by the DOE Project Plan, calculations were performed to estimate for these streams the additional dose presented by constituents in irradiated uranium over that of unirradiated uranium. A fractional dose calculation with a result of <0.1 indicates that the additional dose presented by the RU constituents is less than 10% of the dose expected from doing similar work with uncontaminated weapons-grade uranium. RU streams characterized by a dose fraction of <0.1 were deemed *de minimis* in accordance with the definition established by DOE for the Recycled Uranium Mass Balance Project. For those streams, the radiation-protection measures in place for the presence of uranium are considered adequate for worker protection.

The highly enriched RU from Savannah River and Idaho in the form of uranyl nitrate and uranium oxide was processed at the Y-12 Complex and shipped to Savannah River as highly enriched uranium (HEU) metal for fabrication of production reactor fuel. The primary focus of this document is on the facilities and processes that had the potential for concentrating the RU constituents, relative to the uranium flow, and so presented the greatest potential for increased worker exposure.

Five shipments of slightly enriched RU (0.74% ²³⁵U) were received from SRS; however, they were transferred to Fernald within one day to one month of receipt. Since the material was not repackaged, it is not considered to contribute to an increase in personnel exposure.

Slightly depleted RU oxide was received by the Y-12 Complex from ORGDP, Hanford, and PGDP (including fluorination tower ash from PGDP). Documentation and discussion with many individuals who worked at the Y-12 Complex from the 1950s onward indicated that the plant did not have the need for nor the capability of chemically processing this material. Therefore, it is assumed this material was sent to the plant for storage prior to burial or further disposition to other Oak Ridge Operations sites; most of the ash was returned to ORGDP and PDGP. Since these materials were apparently not processed or handled directly at the Y-12 Complex, they are not at this time considered to be potential sources of increased personnel exposure or significant environmental release. Further analysis may be warranted in the future if these materials are determined to have been processed at the Y-12 Complex.

¹ Slightly depleted RU recovered from production reactor spent fuel is generally in the range of 0.6 to 0.71% enrichment.

Depleted uranium metal primarily from Fernald, produced from gaseous diffusion plant tails, has been used extensively in weapons and defense programs at Idaho, Rocky Flats, the Y-12 Complex, and other sites. Identical material received at Idaho was analyzed in the Report on Mass Balance at the Specific Manufacturing Capability Project where it was determined that the fractional dose resulting from the RU constituents is less than 10% of that of the uranium itself. The ORGDP Mass Balance Report also confirms very low levels of transuranics and Tc in the tails streams. Processing of this material in a manner that concentrated the RU constituents was not performed at the Y-12 Complex; rather, the material was fabricated as is into an end-use form. For this reason, and in accordance with the DOE Project Plan, this depleted uranium metal stream was excluded from further consideration.

III. RECYCLED URANIUM AT THE Y-12 COMPLEX

For purposes of the DOE recycled uranium mass balance project, RU has been defined as any uranium that has been irradiated in a reactor and, as a result, contains TRU material (e.g., Pu and Np), fission products (e.g., Tc), and reactor-generated uranium products (^{236}U). The methodology applied in this Y-12 Complex project for identifying the flow of RU materials includes the criteria of (1) the source site, (2) the isotopic constituents, and (3) the wt-% assays of the material. Sites identified as RU source sites are the U.S. government facilities that operated production reactors and/or used chemical separation processes to extract uranium from irradiated fuel. Primary source sites are SRS, ICPP, and Hanford. The majority of Y-12 Complex transfers with SRS and ICPP have involved RU (although significant quantities of fresh fuel and sweetener² were also shipped to Savannah River). Secondary source sites providing RU materials to the Y-12 Complex are ORGDP and PGDP. The project identified and reviewed RU streams at the Y-12 Complex from the initial introduction of RU into the plant in 1953 until March 31, 1999.

Receipts

RU was received at the Y-12 Complex from three primary source sites:

- ***receipts of 125,161 kg of highly enriched RU*** as UN solution or U-Al ingots from SRS; this material was processed in the plant's 9212 and/or 9206 facilities,
- ***receipts of 25,696 kg of highly enriched RU*** as UN solution or oxide from ICPP; this material was processed in the plant's 9212 and/or 9206 facilities.
- ***receipts of 1,502 kg of slightly depleted RU*** as oxide from Hanford; the assay associated with this material indicates that it was slightly depleted uranium (DU); this material is believed to have been disposed of on the Oak Ridge Reservation without any processing in Y-12 Complex facilities.

The Y-12 Complex also received RU from the following secondary sites:

² HEU used to blend with recycled uranium fuel feed to increase its enrichment is referred to as "sweetener."

- *receipts of 192,836 kg of slightly depleted RU* from ORGDP; this material is believed to have been stored at the Y-12 Complex temporarily and returned to ORGDP and
- *receipts of 38,423 kg of slightly depleted RU* as fluorination tower ash from PGDP; this material is believed to have been disposed of on the Oak Ridge Reservation or returned to PGDP without any processing in Y-12 Complex facilities.

Shipments

RU streams exited the Y-12 Complex via:

- *shipments of 120,384 kg of highly enriched RU* as metal product to SRS,
- *shipments of 29,614 kg of RU* as slightly depleted fluorination tower ash to PGDP (this material was apparently the ash that had been shipped from PGDP to the Y-12 Complex and stored at the plant), and
- *shipments of 192,836 kg of slightly depleted RU* to ORGDP (these represent the return of material to ORGDP).

Inventory

As of March 31, 1999, approximately 13 MT of highly enriched RU remained in the Y-12 Complex inventory.

Summary

The estimated mass balance for highly enriched RU, which is of most concern for worker exposure and is the primary focus of this project, is summarized in Table ES-1. A discrepancy in the mass balance between receipts and shipments (plus inventory and waste) reflects an inability to precisely distinguish between RU and non-RU shipments and receipts involving the Y-12 Complex and Savannah River. Shipments of fresh fuel (non-RU) and sweetener (also non-RU) were made from the Y-12 Complex to Savannah River along with RU shipments. The only way to distinguish between these RU and non-RU streams using available records is by enrichment level. Shipments of $\leq 90\%$ enrichment were assumed to be RU. Shipments of $> 90\%$ enrichment were assumed to be non-RU fresh fuel or sweetener. This methodology using enrichment level to distinguish between RU and non-RU results in good estimates of RU flows that are reasonably consistent with Savannah River estimates. Although this is the best available means of distinguishing RU streams, this method does leave a difference of approximately 17.3 MTU between receipts and shipments.

Slightly depleted RU streams received by the Y-12 Complex from ORGDP and PGDP are believed to have been returned to the shipping site or disposed of as waste on the Oak Ridge Reservation. No evidence of Y-12 Complex processing of this material was identified in the historical records reviewed by the Project Team.

Table ES-1 Estimated Mass Balance for Highly Enriched RU

	RU Received kg U	RU Shipped kg U
Savannah River	125,161	120,384
ICPP	25,696	0
TOTAL	150,857	120,384
Total RU Shipped		120,384
RU Inventory (as of 3/31/99)		13,082
Estimated RU Waste		~100
TOTAL	150,857	133,566
Difference*		~17,300

* This difference is due to the inability to precisely distinguish between RU and non-RU shipments.

IV. CONSTITUENTS (PU, NP, AND TC) IN RU

The overall mass balance for highly enriched RU and constituent flow through the Y-12 Complex is summarized in Table ES-2. This table compiles quantities of each constituent based upon the estimating logic presented in Chapter 5.

Table ES-2 Overall Mass Balance for Y-12 Complex Highly Enriched RU

	Receipts	Shipments	Inventory	Waste	Difference
RU (kg U)	150,857	120,384	13,082	~100	~17,300
Pu (g)	0.051	0.033	0.002	~0.01	~0
Np (g)	3,666	1,073	121	270	2,200 (-300)*
Tc (g)	14,499	12,279	1,365	3,200	-2,345 (335) [†]

* The Np difference is -300 g if it is assumed that the reported 1.75 Ci (2,500 g) Np was buried in the Bear Creek Burial Grounds as solid waste or shipped off site to another DOE facility.

[†] The Tc difference is 340 g if it is assumed that most Tc found in the southeast S-3 Pond came from ORGDP and that Tc is not included in receipts.

Based upon Y-12 Complex records of highly enriched RU receipts and shipments, material remaining in inventory, and determinations regarding quantities in disposal, there remain no more than trace quantities of Pu not accounted for.

In contrast, the overall mass balance cannot account for 2,200 g of Np. In the historical plant record, reference is made to discharge of 2,500 g (1.75 Ci) of Np to the S-3 Ponds. However, the amount of Np that can be accounted for by sampling and analysis of pond sludge is only 145 g. A similar quantity was found in the WETF sludge. It is known by a few individuals in the plant that an ion exchange column was installed in the uranyl nitrate feed stream to specifically remove Np from the incoming SRS RU for use in another program. The spent or loaded ion exchange columns were removed from the feed line and sent off-site for Np recovery. Since there was little residual uranium contained on the ion exchange resin, this transaction was not listed as an RU transfer and was not placed in the plant uranium accountability record. Assuming that the 2,500 g of Np identified in the waste

management record was indeed separated from the RU stream as suspected and either sent off-site for use elsewhere or buried as a solid waste in the Bear Creek Burial Grounds, the overall mass balance shows 300 g more Np than can be accounted for.

The overall Y-12 Complex mass balance shows 2,345 grams more Tc on the plant site than can be accounted for, based on the mass difference between the uranium feed, product, and waste streams. It should be noted that the normal flow of acid waste from the 9212 and 9206 HEU operations to the S-3 Ponds went first into the NE basin. The flow was then routed by overflow pipe to the NW basin, then to the SW basin, and finally into the SE basin. Under this normal design flow pattern, one would expect to find the greatest concentration of Tc in the NE basin and the least in the SE basin. Sludge analysis, however, shows 179 g of Tc in the NE basin, 184 g in the NW, 89 g in the SW, and 2,680 g in the SE. The apparent discrepancy was explained by a former S-3 Pond manager, who stated that on several occasions Tc liquid waste was discharged directly to the SE basin from 5-gal waste drums received from ORGDP. These Tc residues were removed from the gaseous diffusion cascade from time to time during certain maintenance activities. If it is assumed that essentially all of the Tc in the SE basin came from ORGDP and was not included in the Y-12 Complex RU database, the mass balance difference is 340 g Tc, or 2% of the estimated total receipt.

V. POTENTIAL FLOW PATHS OF RU WITHIN THE Y-12 COMPLEX

The processing of RU at the Y-12 Complex impacted a number of facilities and locations at the plant site. The primary facilities with significant involvement in processing RU were:

- Building 9212, a large uranium processing complex that performed uranium recovery operations on RU materials and produced RU metal product,
- Building 9206, a large uranium processing facility that also performed uranium recovery operations on RU materials and produced RU metal product,
- Building 9720-5, the Y-12 Complex “warehouse,” which received, stored, and shipped uranium materials, including RU,
- S-3 Ponds, four holding ponds for liquid and sludge wastes resulting from processes involving uranium, including both unirradiated and recycled uranium (prior to WETF operation beginning in 1986),
- West End Treatment Facility (WETF), a group of nine tanks/bioreactors for holding and treating Y-12 Complex aqueous nitrate wastes (after the S-3 Ponds were taken out of service) plus four sludge storage tanks, and
- New Hope Pond, a large surface water impoundment designed to capture and retain coal fines and other entrained solids from rainwater and plant secondary wastewaters.

Building 9212 Complex Processes

Building 9212 complex processes involved the following pathways:

- receiving UN solution from ICPP (in safe bottles) and from SRS (in tanker trucks)
- weighing SRS tanker trucks (at Building 9929-1)
- sampling UN solution
- pouring UN solution from ICPP safe bottles into “pour-up” stations for transfer to intermediate storage tanks
- pumping UN solution from SRS tanker trucks to 9212
- UN evaporated and concentrated
- manual filling and loading of UN into safe bottles for transfer to 9206 (in the period after 9206 assumed responsibility for certain recovery operations from 9212)
- ICPP UO_3 received and dissolved to produce UN (in the period after ICPP began sending UO_3 instead of UN)
- purification of UN via solvent extraction (primary and secondary extraction)
- pumping of solvent extraction raffinate to S-3 Ponds
- feeding of solvent extraction raffinate to 9212 bioreactor
- transporting of solvent extraction raffinate to WETF
- denitration of uranyl nitrate hexahydrate (UNH) to UO_3
- maintenance on denitrator or fluid beds
- conversion of UO_3 to uranium tetrafluoride (UF_4) in converted lab muffle furnaces
- removal of dry UF_4 from process
- “bomb” reduction of UF_4 to uranium metal
- sampling, fracturing, and packaging of uranium metal buttons
- salvage operations for uranium-aluminum (U-Al) alloy from SRS
- metal product shipped from Building 9720-5

Building 9206 Processes

Building 9206 processes involved the following pathways:

- UN solution “poured-up” into safe tanks
- U-Al ingots received from SRS at Building 9720-5
- dross and sweepings received
- U-Al ingots (or dross/sweepings) dissolved in NaOH to remove Al; sodium diuranate produced
- sodium diuranate dissolved in nitric acid to produce UN
- UO_3 received and dissolved to form UN
- purification of UN via solvent extraction (primary and secondary extraction)
- isolation and transport of raffinate to 9212
- denitration of UNH to UO_3
- maintenance on denitrator or fluid beds
- conversion of UO_3 to UF_4

- removal of dry UF₄ from process
- “bomb” reduction of UF₄ to uranium metal

Processes Associated with Other Y-12 Complex Facilities

- capping and closure of S-3 Ponds and sludge removal and closure of New Hope Pond
- treatment of nitrate waste at WETF
- storage of RU materials at Building 9720-5

VI. EVALUATION OF ACTIVITIES THAT INVOLVED POTENTIAL WORKER EXPOSURE TO RU CONSTITUENTS

Prior to and during the processing of RU, the Y-12 Complex also operated as a uranium-processing facility. Careful consideration for worker protection was given to the introduction of RU for processing. A criterion for acceptance was based upon DOE/OR-859,³ which in turn was derived from an informal agreement between the Y-12 Complex and SRS. The intent of this criterion was to maintain the relative hazard potential of all non-uranium alpha emitters to less than 7% of the relative hazard potential of uranium.⁴ With this limitation, it was expected that RU could be safely managed by the measures already in place for processing uranium.

The Project Team carefully analyzed and evaluated 36 activities identified as involving potential for worker exposure. The team assigned the following Occupational Exposure Potential (OEP) scores:

- | | |
|----------------------|---------------|
| • No Significant OEP | 8 activities |
| • Low OEP | 1 activity |
| • Moderate OEP | 27 activities |

Most of the potential exposure activities at the Y-12 Complex were found to have a “Moderate” OEP rating as a result of the combined product of a constituent level value of 3 for Savannah River RU or a value of 2 for Idaho RU with a value of 1 or 2 for airborne potential and exposure duration. Certain maintenance activities involving equipment that contained finely divided RU solids were assigned a value of 3 for airborne potential. However, because these types of maintenance activities were not performed very often, the overall OEP was rated “Moderate,” with a cumulative score of 9.

In no instance did any identified activity involve a combination of airborne potential, constituent level, and exposure duration that produced an OEP score in the “High” range. Although some activities presented moderate OEP scores, the average derived air concentrations (DAC) for the areas associated with RU was on the order of only 3% of the plant action level (PAL).

The methodology established for the DOE Mass Balance Project considered ²³⁶U an unmonitored isotope, along with Pu, Np, and Tc. In fact, ²³⁶U is generally indistinguishable from other uranium isotopes; it has the same chemical behavior and the same dose

³ Egli, D. et al., *The Report of the Joint Task Force on Uranium Recycle Materials Processing*, 1985.

⁴ Vath and Duerksen, *Criteria for Acceptance and Technical Assessment for Acceptance of Enriched Uranium at the Y-12 Plant*, April 25, 1996.

consequences as can be seen by comparing uranium DAC values. Monitoring, both in the field and through bioassay, accounts for its presence and correctly assigns dose or risk. Other constituents, such as plutonium, are fundamentally different in that they do not have the same chemical behavior or risk. Their presence could alter the intrinsic risk of handling recycled uranium. Because ^{236}U was monitored at the Y-12 Complex, the analysis presented here, which used the DOE Mass Balance Project *de minimis* calculation methodology, estimates the Occupational Exposure Potential (the implied hazard) to be higher than it actually is. A calculation that considers the non-uranium, potentially unmonitored component would at times lead to the conclusion of “No Significant Occupational Exposure Potential” when ^{236}U is more appropriately considered.

VII. EVALUATION OF PROCESSES OR FACILITIES THAT INVOLVED POTENTIAL ENVIRONMENTAL RELEASES

Various sources that documented the potential environmental impact of RU components from the Y-12 Complex and the Oak Ridge Reservation were identified and reviewed by the Project Team. These reports are summarized in Chapter 2.

Solvent raffinate streams from Building 9212 and 9206 extraction systems—as well as condensed acid streams from the various UN solution evaporators and denitrators—were ultimately discharged to the unlined S-3 Ponds. Chemical analysis of the S-3 Pond sludge indicated the presence of 3,140 g of Tc, 145 g of Np, and <0.1 g of Pu. The S-3 Ponds were capped in 1986, with the sludge left in place under EPA oversight. Uranium has been detected in groundwater monitoring wells around the S-3 Ponds. Therefore, one can infer that RU constituents also leached into the nearby environment from the ponds. Data from other locations, such as the WETF and New Hope Pond, were analyzed and indicated these sites have no significant potential for environmental releases.

VIII. CONCLUSIONS

Potential Personnel Exposure

Although the Project Team identified 36 activities as having potential for worker exposure, in no instance did any identified activity produce an OEP score in the “High” range. As a result, the potential for worker exposure to TRU elements and fission products at the Y-12 Complex is considered low to moderate.

Early in its existence, the Y-12 Complex implemented a worker protection program that included worker radiological protection (see Section 2.7). This program incorporated such elements as personnel protective equipment, personnel monitoring, environmental monitoring, work location surveys, work-time limits on jobs with penetrating radiation, excretion rate limits, periodic examinations of personnel, and Plant Action Level limits. The inhalation of radioactive materials was recognized as the most important source of possible exposure at the Y-12 Complex. Consequently, administrative controls were primarily designed to guard against associated hazards.

Worker protection measures in place at the Y-12 Complex likely provided substantial mitigation to the risks introduced by the activities rated as moderate to low in OEP.

However, dose assessment studies may be warranted as a follow-on activity to provide a more detailed assessment of worker exposure.

Potential Environmental Releases

Soil and groundwater around the Y-12 Complex is contaminated with various radionuclides as a direct result of the nature of the Y-12 Complex work and past disposal practices. However, the quantities of RU constituents in and around the plant are very small and pose no threat to the immediate environment or the surrounding communities. A clear understanding of the contamination exists, and ongoing environmental programs continue to verify this conclusion. The report of the joint task force assembled by DOE in 1985 to study past and (then) current practices related to the processing of RU reflected similar conclusions. The task force did not disclose any instance at the Y-12 Complex in which the environment was jeopardized or compromised.

An Oak Ridge Dose Reconstruction Project was initiated in 1994 as follow-up to the Oak Ridge Dose Reconstruction Feasibility Study, which recommended a closer examination of past uranium emissions and potential resulting exposures. The Task 6 component of the project involved further evaluation of Oak Ridge uranium operations and effluent monitoring records to determine if uranium releases from the Oak Ridge Reservation likely resulted in off-site doses that warranted further study. The results were documented in the July 1999 Task 6 report. The Task 6 team concluded that earlier estimates of uranium releases had been underestimated. However, based on the decision guidelines from the Oak Ridge Health Agreement Steering Panel, the Task 6 team concluded that while Y-12 Complex uranium releases are candidates for further study, they are not high-priority candidates.

The Task 7 component of the project involved performing qualitative and quantitative screening of various materials of concern at the Y-12 Complex and the other DOE Oak Ridge sites. Materials screened included Np and Tc. Results were reported in the Task 7 report. Based on the analysis of data, the Task 7 team determined that Np did not warrant further study. Although Tc was identified as one of the potential candidates for further study, it was not determined to be a high-priority candidate.

These analyses, along with other information on environmental consequences from Y-12 Complex operations, identify candidate environmental issues for additional study. However, candidate issues related to the processing of RU have not been determined to be high-priority candidates for further study.